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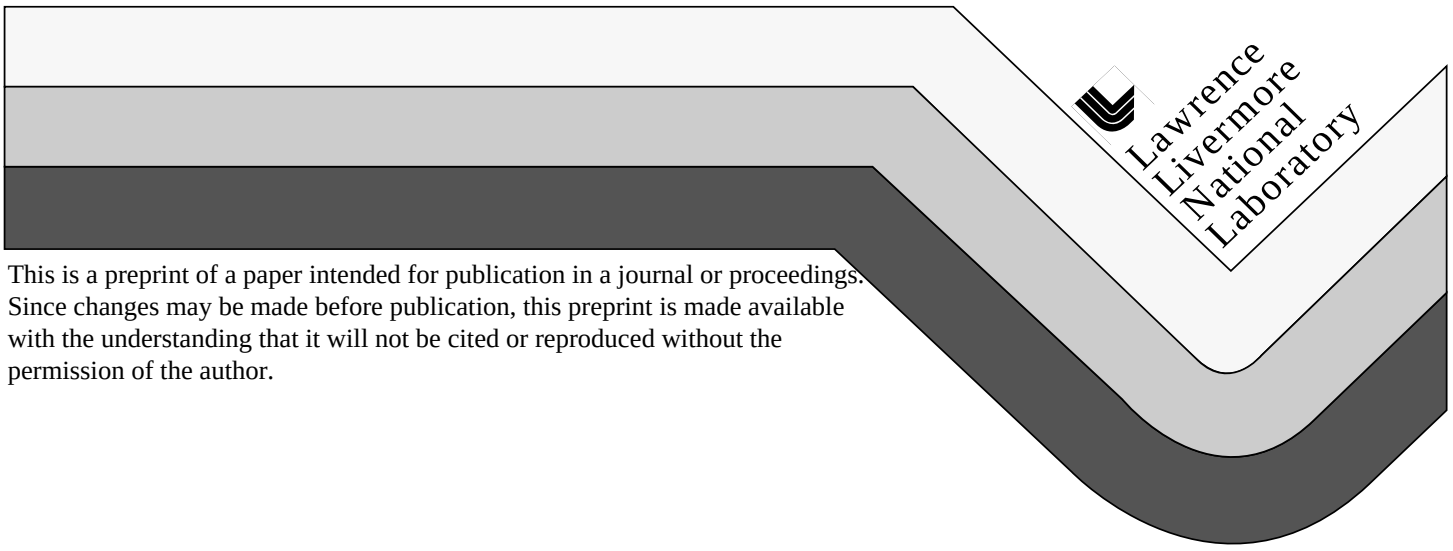

Tunable laser action at 4.0 microns from Fe:ZnSe

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Abstract: The spectroscopic properties of Fe:ZnSe were investigated and 4 micron lasing action was demonstrated for the first time at sub-ambient temperatures. Using an Er:YAG laser as a pump source, the Fe:ZnSe laser produced output energies of 8 μJ/pulse with pulse lengths of 48 μs at a repetition rate of 100 Hz. The output wavelength varied with temperature from 3.98 μm at 14 K to 4.54 μm at 180 K. A slope efficiency of 6.7% at 130 K was obtained.

OCIS codes: (140.5680) Rare earth and transition metal solid-state lasers; (140.3070) Infrared and far-infrared lasers; (140.3600) Lasers, tunable; (140.3380) Laser materials

Introduction

Tunable solid state lasers have been shown to be versatile tools in science and industry, particularly in the areas of remote sensing, military countermeasures, and the detection of organic and inorganic compounds. The current lack of direct, widely tunable 3-5 μm solid state lasers is a strong driver for pursuing new classes of infrared laser materials. The recent demonstration of lasing action in Fe:ZnSe represents the discovery of a new widely tunable solid state laser material which has the potential to operate over a 700 nm wavelength range.

The success of Cr^{2+}:ZnSe as a solid state laser material has motivated much interest in investigating other transition metal-doped chalcogenides [1-3]. The previous work by DeLoach et al. in 1996 suggested Fe^{2+} as a probable candidate for infrared laser action. The energy level diagrams and level designations for Cr^{2+} and Fe^{2+} in a tetrahedral field are shown in Fig. 1. The Cr^{2+} ion has four electrons in its d shell while the Fe^{2+} ion has a complementary electronic structure due to its four holes. This leads to an inversion of the energy levels [4]. In both ions, the crystal field will effectively lower the free-ion symmetry and thus allow d shell to d shell

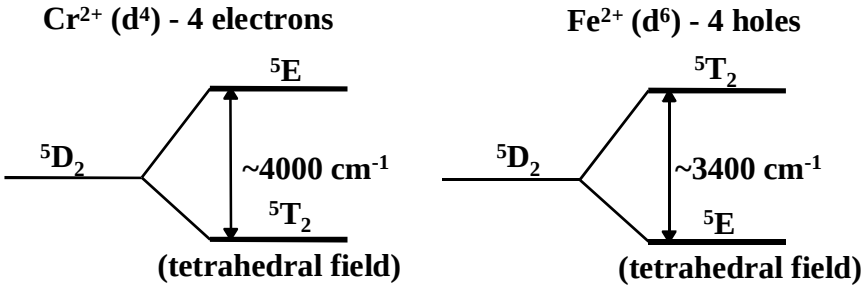

Fig. 1. Energy level diagrams for Cr^{2+} and Fe^{2+} in a tetrahedral field.

transitions. The magnitude of the ^5E to $^5T_2$ splitting for Fe^{2+} in a tetrahedral configuration is estimated to be around 3400 cm^{-1}. Only spin allowed transitions occur between the ^5E and $^5T_2$ levels and therefore we do not expect significant excited state absorption losses (ESA). Since the lasing transition only

involves valence electrons the broadening due to phonons in the absorption and emission bands is expected to provide wide tunability.

II-VI compounds have a tetrahedral coordination and have been previously used as hosts in several infrared laser materials such as Cr:ZnSe, Cr:ZnS, and Cr:CdSe. The II-VI compounds are suitable infrared hosts for Fe^{2+} since they transmit beyond 10 μm and have phonon energies ~300 cm^{-1}. The low phonon energies will render nonradiative processes multi-phonon and therefore we expect radiative processes to dominate below a certain temperature. In addition, since the Fe^{2+} ionic radius is similar to Zn^{2+} and Cd^{2+} this allows good substitutional incorporation into these host materials. The Bridgman growth and diffusion doping methods produced crystals of Fe^{2+}:ZnS and Fe^{2+}:ZnSe with reasonable optical quality.

The ZnSe and ZnS host materials have been shown to have robust thermomechanical properties [2]. As shown in Table 1, ZnS and ZnSe have higher thermal conductivities (κ) and thermal shock resistance parameters (R_T) than YAG, but a factor of 5-10 larger dn/dT. As a consequence, thermal lensing will probably need to be compensated in cavity designs of medium to high power laser systems.

Table1. Thermomechanical properties of ZnS, ZnSe, and YAG [2]

Material	κ (W/m°C)	R_T (W/$m^{1/2}$)	$\frac{dn}{dT}$ (10^{-6}/°C)
ZnS	**17**	**7.1**	**+46**
ZnSe	**18**	**5.3**	**+70**
YAG	**10**	**4.6**	**+8.9**

Spectroscopy

We conducted a preliminary spectroscopic study of the Fe-doped crystals to assess their feasibility as laser materials. The absorption spectra shown in Fig. 2 were measured using an Air Products

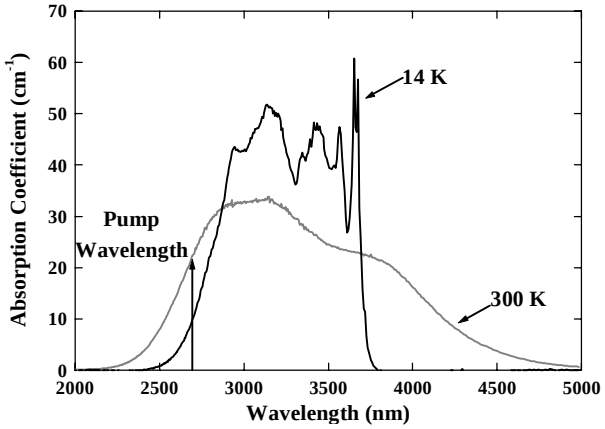

Fig. 2. Absorption Spectra for Bridgman grown Fe^{2+}:ZnSe at 14 K and room temperature.

Displex helium gas cryogenic system and a computer controlled Perkin-Elmer 983 Infrared Spectrophotometer. These spectra suggested that an available Er:YAG laser operating at 2698 nm would make a suitable pump source for the luminescence and laser experiments. In the 14 K spectrum,

the resonance structure in the absorption band is revealed. Also, the wavelength broadening of the absorption with temperature, as is common in vibronic materials, widens the spectrum out to 5 μm at 300 K. Fig. 3 shows an uncalibrated emission spectrum for Fe:ZnSe measured with an Er:YAG laser as the excitation source. The experiment employed a helium gas cryogenic system, a 1 meter 300 groove/mm McPherson monochrometer, an EG&G Judson InSb liquid nitrogen cooled detector, and a lock-in amplifier. The emission band exhibits a broad feature with a FWHM of 400 nm leading to the potential for tunability from 3.7 to 4.5 μm. The fluorescence lifetime vs. temperature plot in Fig. 4 shows observable emission out to 200 K.

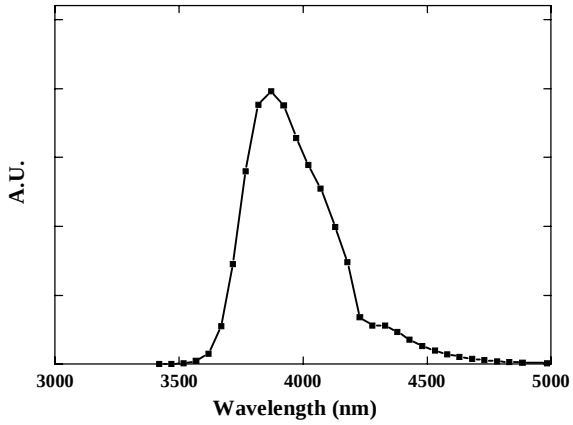

Fig. 3. Uncalibrated spontaneous emission spectrum for Fe^{2+}:ZnSe at 77 K.

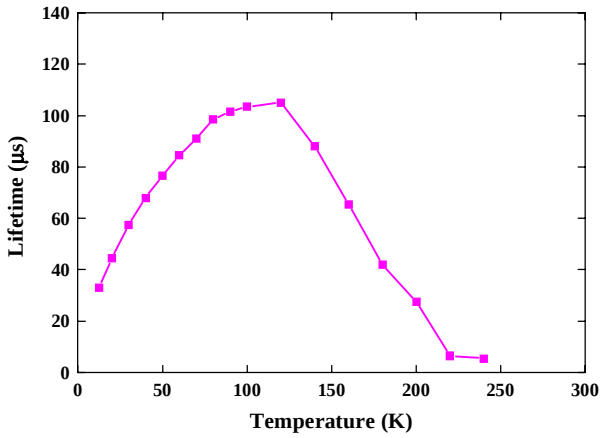

Fig. 4. Fluorescence lifetime vs. temperature for Fe^{2+}:ZnSe.

The lifetimes ranged from 5 μs to 110 μs for temperatures less than 220 K thus alleviating any particularly demanding requirements on the pump pulselengths.

Laser Experiments

A confocal laser cavity using a pair of 10 cm mirrors was built around a cryogenically cooled Fe:ZnSe sample. A diagram of the experimental laser system is shown in Fig. 5.

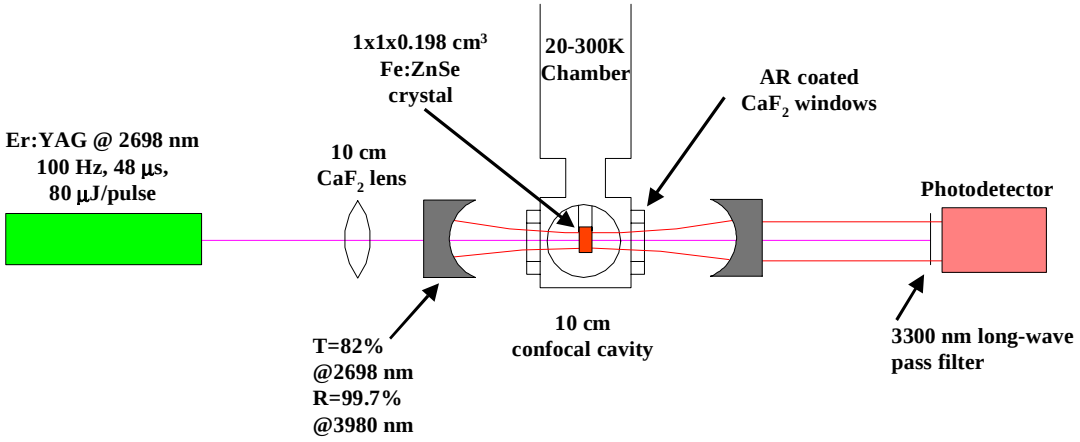

Fig. 5. Fe^{2+}:ZnSe laser system.

The Fe:ZnSe crystal was cooled by a helium gas cryogenic system and a liquid nitrogen cooled InSb detector was used to detect the laser signal. The cryogenically cooled Fe:ZnSe sample was end-pumped by the Er:YAG laser which operated at a wavelength of 2698 nm and delivered 48 μs pulses at a repetition rate of 100 Hz. The Er:YAG pump laser had a maximum output energy of ~80 μJ/pulse. The mirrors of the cavity were identical and the waist size of the cavity was 250 μm within the uncoated Fe:ZnSe crystal. Lasing of Fe:ZnSe was observed from 15 K to 180 K. The Fe:ZnSe laser's operating parameters mimicked those of the Er:YAG pump laser. The Fe:ZnSe laser produced 48 μs pulses at a repetition rate of 100 Hz with a peak output energy of ~8 μJ/pulse.

The slope efficiency data was taken with the same basic experimental setup as in Fig 5, except that the InSb detector was replaced by a Molectron J3-09 joulemeter and a second Molectron J3-09 joulemeter was used to reference the incident pump energy. The output energy of the Fe:ZnSe laser was measured as a function of 2.698 μm incident pump energy. This data is shown in Fig. 6. For a 0.3% output coupling, Table 2 lists slope efficiencies, thresholds, and upper bounds for the passive losses for the Fe:ZnSe laser.

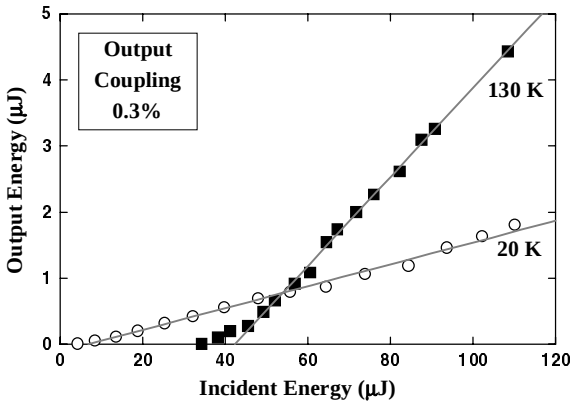

Fig. 6. Fe^{2+}:ZnSe laser output energy vs. 2.698 μm incident pump energy.

Table 2. Fe^{2+}:ZnSe laser operating parameters

Temperature (K)	Slope Efficiency	Threshold	Passive Losses (upper bound)
20	**1.6 %**	**5 μJ**	**12.4 %**
130	**6.7%**	**41 μJ**	**2.4 %**

The low values for the slope efficiencies can be improved by optimizing the output coupling in addition to reducing the bulk loss in the crystal and AR coating the surfaces of the crystal.

The output energy and wavelength of the Fe:ZnSe laser was observed to change with temperature as reflected in Fig. 7. The output energy measurements were made using the same experimental arrangement as for the slope efficiency measurements, except that the incident energy from the Er:YAG pump laser was kept constant and the temperature of the Fe:ZnSe crystal was allowed to change in discrete steps. The output wavelength data was collected using a 1/3 meter, 150 groove/mm CVI Instruments Digikrom 240 computer controlled monochrometer, a Labview data acquisition program, and a liquid nitrogen cooled InSb detector. This equipment replaced the photodetector on the right side of Fig. 5. Fig. 7 shows that the output wavelength of the Fe:ZnSe laser increases

monotonically with temperature. The Fe:ZnSe laser is thus temperature tunable over the range of at least 3980 nm to 4540 nm.

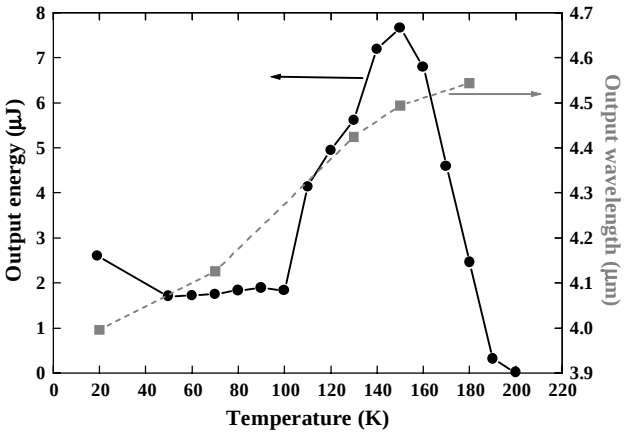

Fig.7. Output energy and wavelength vs. temperature for the Fe^{2+}:ZnSe laser.

The low observed output energy between 40 K and 90 K is believed to be due to a strong CO_2 absorption near 4230 nm which could have strongly attenuated the Fe:ZnSe laser output. No effort at that time was made to purge the CO_2 in the region between the laser output and the detector, but actions to reduce the effects of the CO_2 absorption in the atmosphere are planned for the future.

Conclusion and Future Directions

Fe^{2+} in ZnSe has been demonstrated as an infrared laser with the potential for tunability. For the future, we plan tuning experiments with the Fe:ZnSe laser as well as Q-switching and possibly mode locking. As can be seen in Fig. 8, various transition metals in oxide and fluoride hosts span one region of the infrared, Fe^{2+} and Cr^{2+} in II-VI hosts span another, and maybe other transition metals have the potential to encompass even more of the infrared.

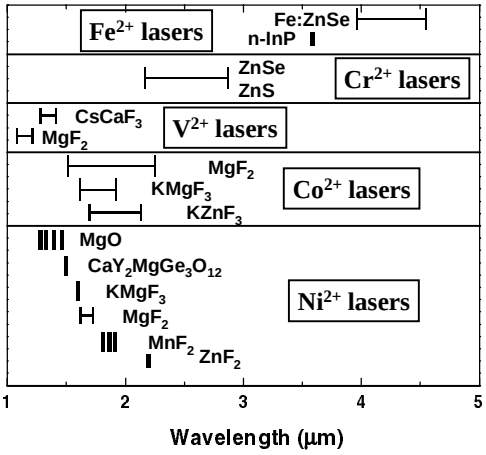

Fig. 8. Common transition metal infrared laser materials.

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